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Observation of competing order in a high-T_c superconductor using femtosecond optical pulses

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In the Bardeen-Cooper-Schrieffer (BCS) theory of superconductivity, which describes the mechanism of conventional superconductivity for metals, electrons form Cooper pairs via interactions mediated by the vibrations of the crystal lattice. For the high-temperature superconductors, another possibility exists, namely, Cooper pairing via antiferromagnetic spin fluctuations. Indeed, a full-fledged antiferromagnetic order, out of which such antiferromagnetic fluctuations emerge, can also compete with superconductivity as the dominant ground state resulting in phase coexistence.

The coexistence of antiferromagnetic ordering with superconductivity has been observed in single- or double-layer systems in the presence of a magnetic field via neutron scattering, or in five-layered systems in zero field using nuclear magnetic resonance. However, it is unclear from these measurements how the emergence of antiferromagnetic order affects the quasiparticle (QP) excitations, which determine the material's optical and electronic response.

In recent years, femtosecond time-resolved spectroscopy has been recognized as a powerful bulk technique to study temperature-dependent changes of the low-lying electronic structure of superconductors and other strongly correlated electron materials. It provides a new avenue, namely the time domain, for understanding the QP excitations of a material. We present time-resolved studies of photoexcited QP dynamics in the high-T_c superconductor Tl-2223 (T_c=117K). We observe that its pristine superconducting state (40K<T<T_c) subsequently evolves into a coexistence phase as evidenced by a strong modification of the gap dynamics below 40K.

In femtosecond pump-probe experiments, a femtosecond laser pump pulse excites quasiparticles. These high-energy quasiparticles rapidly thermalize (within tens of femtoseconds) via electron-electron collisions, reaching states near the Fermi energy. The subsequent relaxation dynamics are strongly affected by the low-energy electronic structure in these materials. The dynamics are extracted either by time-resolved measurements of the photoinduced changes in reflectivity ($\Delta R/R$) or transmission ($\Delta T/T$) at optical frequencies, or by directly measuring conductivity dynamics, with the probe wavelength in the terahertz (far-infrared) range.

Figure 1 shows the crystal structure of Tl-2223, with three CuO_2 planes in a unit cell. Figure 2 shows the time dependence of the photoinduced signal of Tl-2223 in the vicinity of T_c. At high temperatures the signal is characterized by a negative $\Delta R/R$ transient which relaxes within τ_n~0.5 ps with τ_n decreasing slightly as temperature is increased to

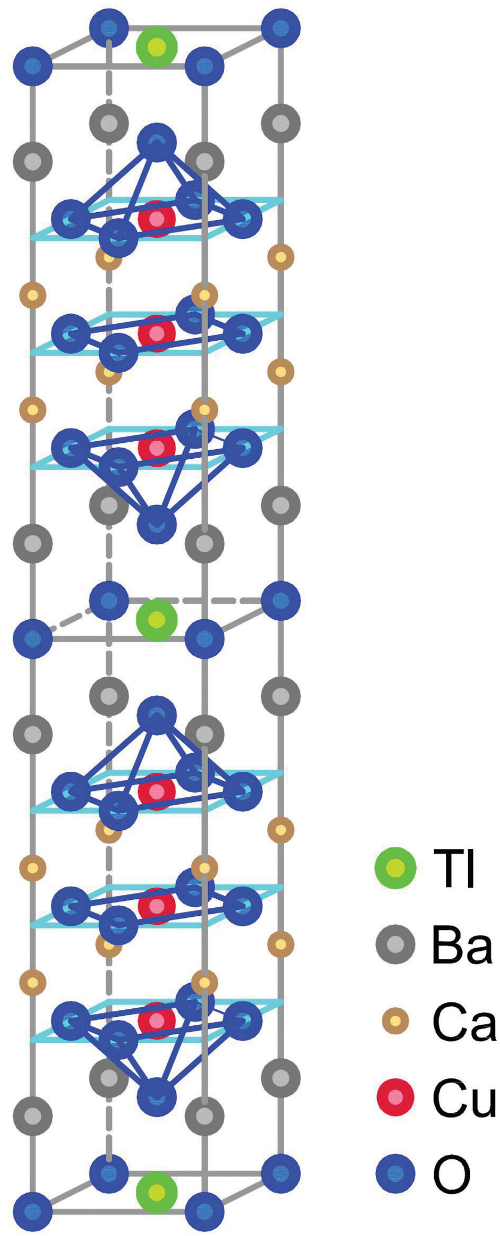

Figure 1. Crystal structure of Tl-2223, with three CuO_2 planes per unit cell. Notice that the two outer CuO_2 planes has a pyramidal coordination with an apical oxygen, while the inner plane has a square coordination with no apical oxygen.

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300K (Fig. 2a).

Below T_c, we observe the onset of a positive $\Delta R/R$ with a relaxation time (τ_{SC}) of a few picoseconds (Fig. 2b). Surprisingly, below ~40K, $\Delta R/R$ first goes positive, relaxes to zero with a lifetime τ_{SC}, *then crosses zero* and goes negative, before relaxing back to equilibrium over a time scale of a few hundred picoseconds (τ_φ) (Fig. 2c).

We ascribe the short-decay positive signal to the reformation of superconducting order following photoexcitation, and the long-decay negative signal to the development of a new competing order other than superconductivity.

We use the Rothwarf-Taylor (RT) model to explain our data. It is a phenomenological model used to describe the relaxation of photoexcited superconductors, where the presence of a gap in the electronic density-of-states gives rise to a relaxation bottleneck for carrier relaxation. When two quasiparticles with energies $\geq\Delta$, where Δ is the superconducting gap magnitude, recombine, a high-frequency phonon (HFP) with energy $\omega\geq 2\Delta$ is created. HFPs released during quasiparticle recombination that remain in the excitation volume can re-break Cooper pairs; hence they act as a bottleneck for quasiparticle recombination, and superconductivity recovery is governed by the decay of the HFP population.

We first use the RT model to analyze the competing component below T_φ, as well as the superconducting component in the range $T_\varphi<T<T_c$, where only one order parameter exists in the system, namely *d*-wave superconductivity. We have shown[2] that the relaxation dynamics of the competing order and the pure superconducting order can be explained by the RT model, i.e. the presence of a gap in the density of states, resulting in a relaxation bottleneck. The fits using the RT model to our data are shown in Figures 3a and 3b.

We also notice from Figure 3b that below T_φ, the fitted values of the relaxation time (dashed line) underestimate the experimental values. This deviation must be due to the presence of the new order, which competes with the superconducting order and depresses the superconducting gap. In the Ginzburg-Landau theory[1], the coupling between the competing and superconducting order parameters causes the latter to be suppressed. Hence the superconducting energy gap Δ_{SC} decreases below its BCS value, as shown in Fig. 3d. We infer that, below T_φ, the increase of the experimental relaxation time $\tau_{SC}^{\exp}(T)$, over its BCS value $\tau_{SC}^{pure}(T)$, is due to the suppression of the superconducting gap in this temperature range.

Figure 3c shows the ratio $\frac{\tau_{SC}^{expt}}{\tau_{SC}^{pure}}$ (circles), and the fit using the RT model.

Our analysis thus shows that the deviation of our data from the BCS temperature dependence below T_φ is due to the *depression* of the superconducting gap, which is caused by the appearance of the second order. It confirms the *competing* nature of this new order below T_φ. Our data are therefore not inconsistent with the coexistence of antiferromagnetism and superconductivity below ~40K.

Our work is unique compared to other techniques

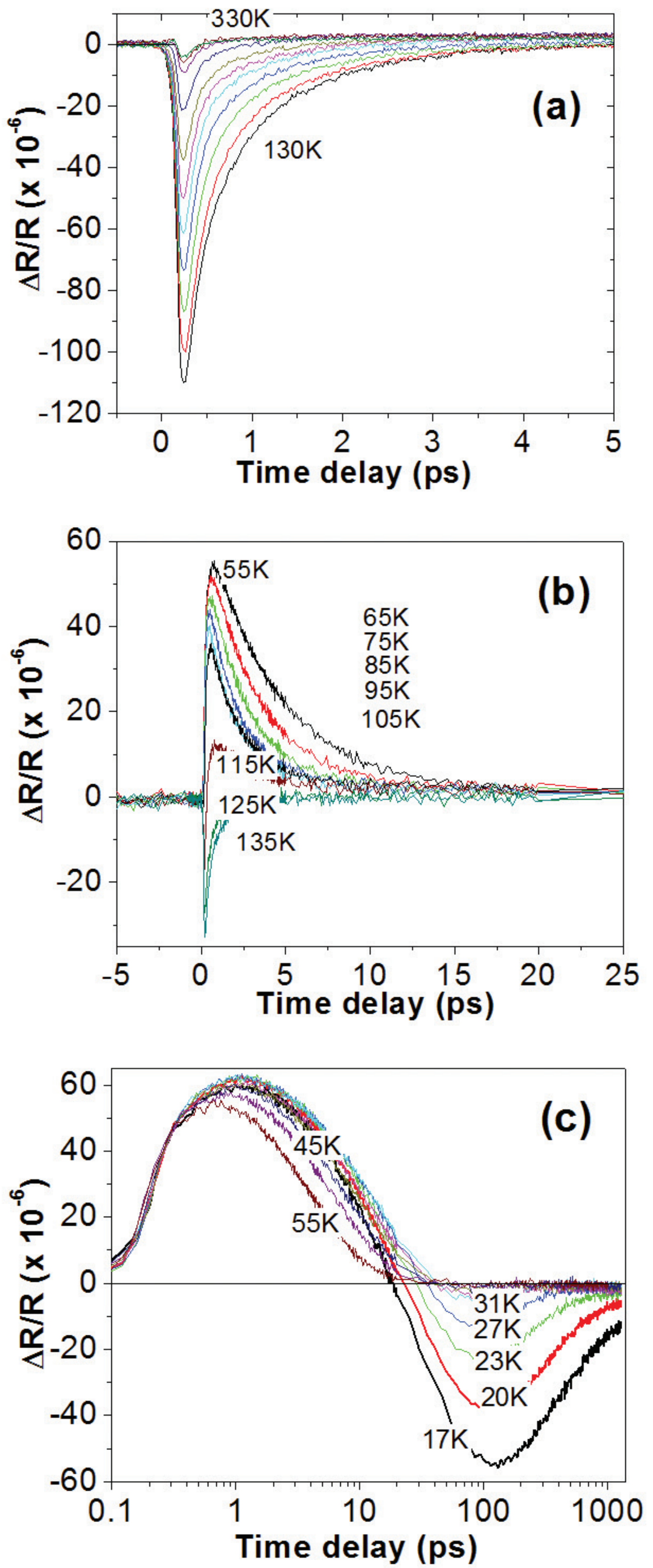

Figure 2. Photoinduced transient reflection $\Delta R/R$ versus time delay between pump and probe pulses, at a series of temperatures through T_φ and T_c. The logarithmic scale is used for the time-axis in the low-temperature region (figure c).

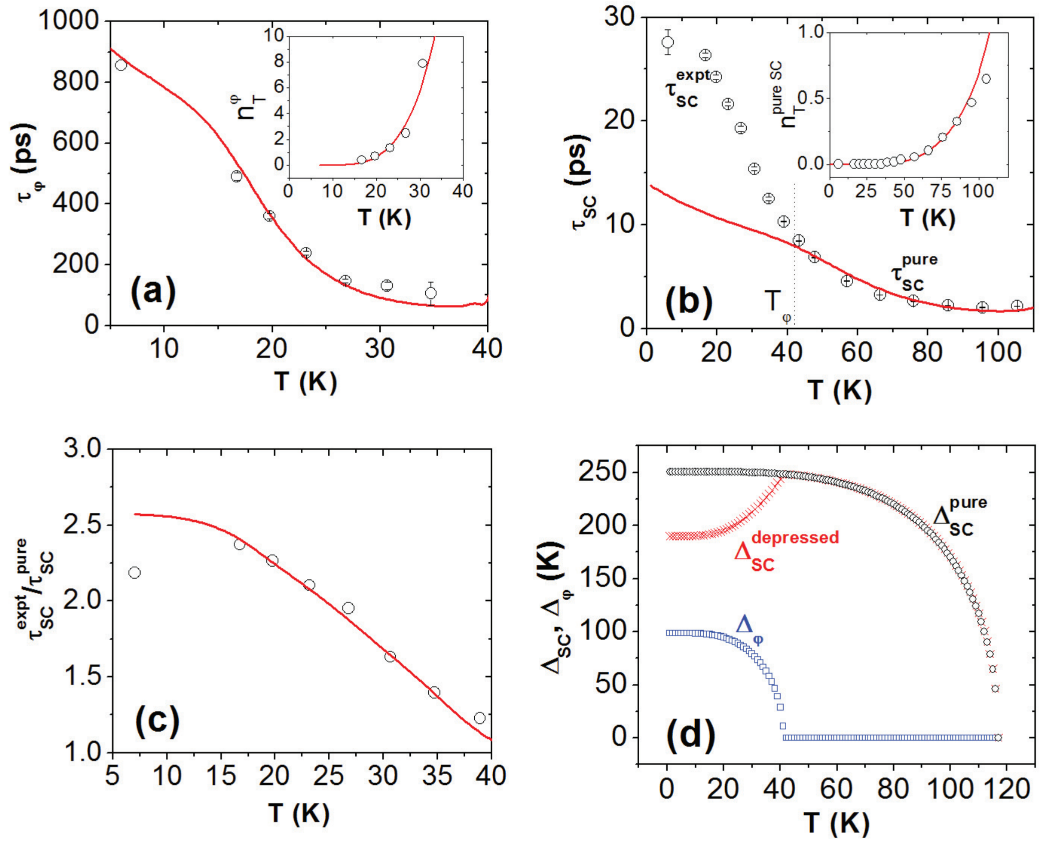

Figure 3. Temperature dependence of relaxation times τ and thermally-excited quasiparticle densities n_T.
a) Relaxation time $C_\varphi(T)$ and thermally excited quasiparticle density $n_T(T)$ of the competing order component. Solid curves are fits using the RT model.
b) Relaxation time $\tau_{SC}(T)$ and thermally excited quasiparticle density $n_T(T)$ of the superconducting component. Solid curves are fits using the RT model. The RT model fit to $\tau_{SC}(T)$ is only for $T_\varphi < T < T_c$ (solid line). The dashed line is the extrapolation of that fit below T_φ.
c) Experimental values and theoretical fit of the $\frac{\tau_{SC}^{expt}}{\tau_{SC}^{pure}}$ ratio.
d) Superconducting gap $\Delta_{SC}(T)$ with and without suppression, and the gap $\Delta_\varphi(T)$ due to the competing order, using the Ginzburg-Landau theory.

in various aspects. First, we see the coexistence phase in zero field, while neutron scattering data on $La_{2-x}Sr_xCuO_4$ and $Nd_{1.85}Ce_{0.15}CuO_4$ sees the emergence of the antiferromagnetic phase only with an externally applied magnetic field. Thus our data are not complicated by the presence of vortex lattice and/or stripe order.

Second, this is the first observation of the coexistence phase using ultrafast spectroscopy, a tabletop setup compared to large facilities required for neutron scattering experiments.

Third, our technique only requires a sample volume of ~10^{-10}cm^3 (due to a small laser spot diameter of 60μm and skin depth in the cuprates of 80nm), which is orders of magnitude smaller than that in neutron scattering (~1cm^3). This makes our technique especially suitable for ultrathin platelet-like samples such as the cuprates, enabling us to probe a much wider class of cuprate superconductors.Ours is the first observation of the coexistence of a competing order with superconductivity in a tri-layered system.

Fourth, we have successfully applied the RT model to systems with more than one gap in the density of states, where we quantified the reduction of the superconducting gap in the presence of a competing order. Though our technique cannot determine whether this new order is magnetic or not, our data clearly show that it competes with superconductivity.

The emergence of this new order opens a QP gap, and our data can be fit excellently with a BCS-like gap, indicating the new order is not inconsistent with a commensurate antiferromagnetic spin-density-wave as revealed in zero-field NMR data on five-layered polycrystalline cuprates.

We ascribe the short-decay positive signal to the reformation of superconducting order following photoexcitation, and the long-decay negative signal to the development of a new competing order other than superconductivity.

We do not exclude the possibility that the competing order can be *d*-density wave order, circulating current order, or charge density wave order. Contrast this coexistence phase at zero field in multi-layered samples with the situation in single-layered cuprates, where at a finite hole doping only a single superconducting phase exists and the competing phase must be induced by an external perturbation such as dc magnetic field.

A possible reason is that for multi-layered cuprates, the competing and superconducting order may nucleate on different planes, with each of their correlation lengths much larger than the interlayer distance, such that the two orders can penetrate into each other even at zero magnetic field. It is precisely the ability of ultrafast spectroscopy to temporally resolve the dynamics of different degrees of freedom that enables us to observe these two orders in the coexistence phase.

Our study once again points to the unique characteristics that high-temperature superconductivity results from the competition between more than one type of order parameter. It provides an insight into the mechanism of strongly correlated superconductivity—the quantum fluctuations around this competing order might be responsible for gluing the electrons into Cooper pairs[2].

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